# High-Field Magnetization of Heavy-Fermion Superconductor $UNi_2Al_2$

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**_Abstract_-We report on the magnetization of the heavy-fermion superconductor $UNi_2Al_3$ measured at 1.5 and 4.2 K in magnetic fields up to 35 T. The results indicate in-plane type of magnetocrystalline anisotropy. At both temperatures, almost linear magnetization curves up to the highest field applied are found. In 35 T, we obtained magnetization values of 0.32 $\mu_B$/f.u. at 1.5 K and 0.30 $\mu_B$/f.u. at 4.2 K.**

## I. Introduction

Antiferromagnetic correlations are thought to play an important role in the formation of the superconducting state of heavy-fermion superconductors [1]. Until 1987, the coexistence of superconductivity and long-range antiferromagnetic order was generally accepted to be excluded. The neutron-scattering experiments of Broholm et al. [2] on the heavy-fermion superconductor $URu_2Si_2$ [3] demonstrated for the first time, that magnetism may persist in the superconducting state. Since that time, long-range magnetic order was reported for several other heavy-fermion superconductors e.g. $UPt_3$ [4],[5] and some $(U,Th)Be_{13}$ compounds. However, in all these cases the uranium moments were found to be very small (of the order of magnitude of $10^{-2}$ $\mu_B$).

The study of the interplay of antiferromagnetism and superconductivity has received a considerable impetus with the discovery of the antiferromagnetic heavy-fermion superconductors $UNi_2Al_3$ ($T_N$ = 4.6 K, $T_c$ = 1 K) [7] and $UPd_2Al_3$ ($T_N$ = 14 K, $T_c$ = 2 K) [8], both crystallizing in an ordered variant of the hexagonal $CaCu_5$-type of structure ($PrNi_2Al_3$ type) [7], [8]. For $UNi_2Al_3$, this structure has been confirmed in ref. 9. In both compounds the presence of heavy quasiparticles is evidenced by moderately enhanced electronic contribution $\gamma$ to the normal state specific heat, which amounts 120 mJ/mol$K^2$ for $UNi_2Al_3$ [7] and to 150 mJ/mol$K^2$ for $UPd_2Al_3$ [8]. In contrast to the heavy-fermion superconductors mentioned above superconductivity and long-range antiferromagnetic order coexist with fairly large ordered uranium 5*f*-moments. By means of neutron diffraction the ordered moment in $UPd_2Al_3$ was determined to be 0.85 $\mu_B$ [10], while from $\mu^+$SR studies an ordered moment of about 0.12 $\mu_B$ is found for $UNi_2Al_3$ [11]. As for the moment direction, the moments are thought to be aligned within the basal plane in $UPd_2Al_3$ [10], while the results for $UNi_2Al_3$ were interpreted in terms of moments parallel to the *c* axis [11]. Measurements performed on single-crystalline $UPd_2Al_3$ confirm the alignment of the moments within the basal plane [12]-[14]. The destruction of the antiferromagnetic state upon application of a magnetic field is reflected in a sharp metamagnetic-like transition at about 18 T [13]. Although some single crystals of $UNi_2Al_3$ have successfully been grown by laser-heated pedestal growth at FOM-ALMOS, University of Amsterdam [15], at present no single crystals of this compound, which exhibit the full range of superconducting properties, are available. This may be partly due to metallurgical problems, which were already mentioned in the original paper [7], but could also originate from slight deviations from the exact stoichiometry, which yield a strong suppression of the superconductivity [16]. Therefore, magnetization measurements on single crystals, which could confirm the *c* axis to be the easy-magnetization axis, are lacking. Furthermore, band-structure calculations by Sticht and Kübler [17] yield moments located within the basal plane for both compounds, $UPd_2Al_3$ and $UNi_2Al_3$. These contradictory results motivated us to perform high-field magnetization measurements in order to shed more light on the type of anisotropy.

## II. Sample preparation and characterisation

A polycrystal of $UNi_2Al_3$ was prepared by arc-melting appropriate amounts of the elements with purities of 3N for uranium, 4N8 for nickel and 5N for aluminium. No further heat treatment was given to the sample. By X-ray diffraction, the proper crystal structure was confirmed with lattice parameters *a* = 520.3 pm and *c* =402.1 pm, which are in good agreement with the parameters in ref. 9. A small amount of a secondary phase (smaller than 3%), most probably $UAl_2$, was estimated to be present from some additional lines found in the X-ray pattern.

## III. Experimental results and discussion

AC-susceptibility measurements showed a relatively broad superconducting transition starting below 0.6 K, which is considerably lower than the transition temperature $T_c$ = 1 K reported in ref. 7 for an annealed sample. As we are dealing

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with an as-cast sample, this difference originates most probably from some inhomogeneity in the composition of the sample. In ref. 7, a considerable increase of the superconducting transition temperatures upon annealing was reported. Furthermore, our sample may be slightly off-stoichiometric, which yields similar effects [16].

Specific-heat measurements were performed between 0.3 and 6 K using the standard relaxation time method. For temperatures below 0.5 K an increase of the specific heat with decreasing temperature is observed, which indicates the onset of superconductivity. The expected drop of the specific heat at lower temperatures was not observed down to the lowest temperature measured and probably will occur at even lower temperatures. A relatively broad maximum around 4 K is connected with the magnetic ordering.

Magnetization measurements in magnetic fields up to 35 T were performed on two kinds of samples: particles of a grain size smaller than 100 $\mu$m (which are assumed to be single crystalline), firstly, free to be oriented by the applied field, and secondly, fixed in random orientations by frozen alcohol. The former measurement (free powder) represents a single-crystal measurement along the easy-magnetization axis, whereas the latter measurement (fixed powder) simulates a measurement on an 'ideal' polycrystal. Comparing both results, information about the type of magnetocrystalline anisotropy can be gained. Furthermore, the free-powder measurement in such high fields gives an indication of the size of the ordered moment.

In Fig. 1, the free- and fixed-powder results at 4.2 K are shown. Almost linear magnetization curves and strong magnetocrystalline anisotropy, yielding a magnetic response of about 0.30 $\mu_B$/f.u. for the free-powder and 0.27 $\mu_B$/f.u. for the fixed-powder samples, in 35 T are found. At 35 T, the ratio of the magnetization value $M_{fix}/M_{free}$ is about 0.9. In order to compare this value with the one expected for the proposed uniaxial anisotropy [11], we have to consider an expected ratio $\chi_{fix}/\chi_{free}$. No metamagnetic transition like in $UPd_2Al_3$ [13] is seen indicating the applied fields are not sufficient to destroy the antiferromagnetic alignment of the moments (i.e. the antiferromagnetic exchange interactions are stronger than the applied field). The susceptibility of fixed powder, which simulates a polycrystal, is described by

$$\chi_{fix} = (\chi_a + \chi_b + \chi_c)/3$$

In a hexagonal lattice, the in-plane susceptibilities are equal, i.e. $\chi_a = \chi_b$. Therefore $\chi_{fix}$ is given by

$$\chi_{fix} = (2\chi_a + \chi_c)/3 \qquad (1)$$

The free-powder magnetization gives the response of the axis, where the highest susceptibility is found (i.e. the easy-magnetization direction). In antiferromagnetic uranium intermetallics, the susceptibility for the direction parallel to the moments is found to be larger than for directions

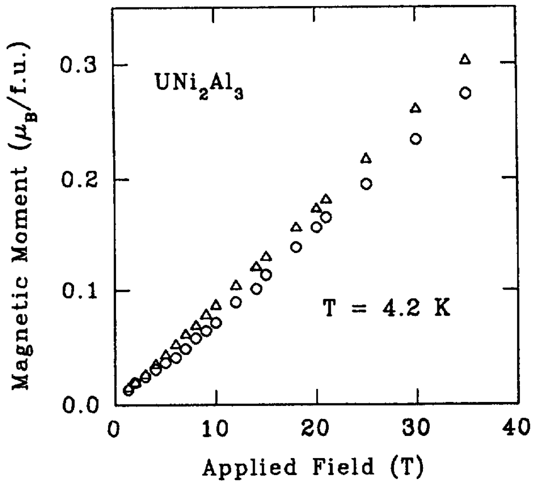


Fig. 1: Field dependence of the magnetization of $UNi_2Al_3$ at 4.2 K obtained on free (△) and fixed powder (○).

perpendicular to the moments. This behaviour has been found for single-crystals of several UTX compounds [18] and also on $UPd_2Al_3$ [12],[13]. Note, that this finding is different from the expectations for a normal Heisenberg antiferromagnet, which yields a perpendicular alignment of the moments to the applied fields. If the *c* axis is the easy-magnetization direction (i.e. $\chi_{free} = \chi_c$), then

$$\frac{\chi_{fix}}{\chi_{free}} = \frac{1}{3} + \frac{2}{3}\left(\frac{\chi_a}{\chi_c}\right).$$

The value of $\chi_{fix}/\chi_{free} = M_{fix}/M_{free}$ of 0.9 found for the magnetization of $UNi_2Al_3$ suggests the perpendicular susceptibility to be 15% smaller than the parallel susceptibility. Comparing this with $UPd_2Al_3$, where the ratio $\chi_\perp/\chi_\parallel \approx 0.5$ [12],[13], it is very unlikely that the *c* axis is the easy-magnetization direction. In contrast, for an easy-plane system (i.e. $\chi_{free} = \chi_a$), which is described by

$$\frac{\chi_{fix}}{\chi_{free}} = \frac{2}{3} + \frac{1}{3}\left(\frac{\chi_c}{\chi_a}\right),$$

$\chi_\perp/\chi_\parallel \approx 0.3$ accounts for the observed $\chi_{fix}/\chi_{free}$ ratio. Assuming a value of $\chi_\perp/\chi_\parallel = \chi_c/\chi_a = 0.5$, which would agree with values found for $UPd_2Al_3$ [12],[13] and for UTX compounds [18] the ratio $\chi_{fix}/\chi_{free}$ should amount 0.83. This value is lower than that one observed, which may in part be due to some remaining texture in the grain, but it can also be connected with the proximity of the ordering temperature. Therefore we checked the results at 1.5 K. In Fig. 2, the result for the free-powder magnetization is shown. In 35 T, a slightly higher magnetization (0.32 $\mu_B$/f.u.) was found than at 4.2 K. In the field range studied, the 1.5 K result confirms the absence of a metamagnetic transition similar to that found in $UPd_2Al_3$ [13]. However, the slight deviation of the magnetization values from linearity toward higher values may be a precursor of a transition to be expected at even higher fields. On the other hand, the present data are not accurate enough to decide whether this deviation is real. Fixed-powder measurements were performed only up to 14 T resulting in a value of $M_{fix}/M_{free}$ of about 0.85 at 14 T, which is close to the expected value of 0.83 for $\chi_c/\chi_a = 0.5$.

## IV. Conclusions

In contradiction to ref. 11, our high-field magnetization results are suggestive of an easy-magnetization direction within the hexagonal basal plane. This finding is supported by the fact that the shortest uranium-uranium distance in $UNi_2Al_3$ is found along the *c* axis, which generally is found to coincide with the direction of the weakest magnetic response [18]. Further support is found in the in-plane alignment of the moments in $UPd_2Al_3$, which crystallizes in the same structure, as reported in ref. 10. Going from Pd to Ni an increase of the *5f*-ligand hybridization (i.e. a decrease of the ordered moment) is expected, whereas a change of the type of magnetocrystalline anisotropy is not very likely.

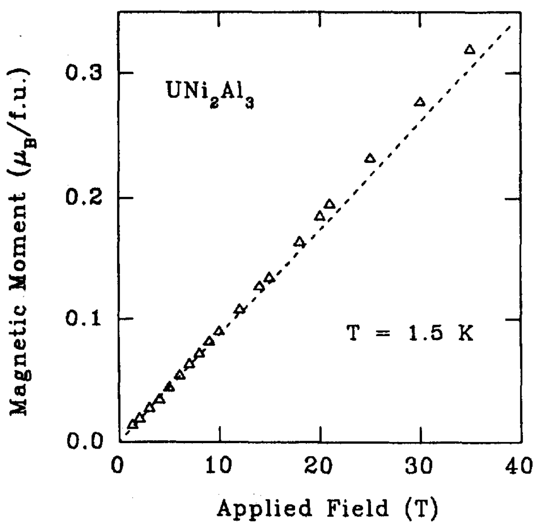


Fig. 2: Field dependence of the magnetization of $UNi_2Al_3$ at 1.5 K measured on free powder. The line shows the deviation from linearity at higher fields.

## References


[1] A. de Visser, J.J.M. Franse, and J. Flouquet, "Interplay and competition in heavy-fermion systems," *J.Magn.Magn.Mater.*, vol. 108, 1992, pp. 15-18.

[2] C. Broholm et al., "Magnetic excitations and ordering in the heavy-electron superconductor $URu_2Si_2$," *Phys.Rev.Lett.*, vol. 58, 1987, pp. 1467-1470.

[3] T.T.M. Palstra et al., "Superconducting and magnetic transitions in the heavy-fermion system $URu_2Si_2$," *Phys.Rev.Lett.*, vol. 55, 1985, pp. 2727-2730.

[4] C. Broholm et al., "Anisotropic temperature dependence of the magnetic-field penetration in superconducting $UPt_3$," *Phys.Rev.Lett.*, vol. 65, 1990, pp. 2062-2065.

[5] G. Aeppli et al., "Magnetic order in $UPt_3$," *Phys.Rev.Lett.*, vol. 60, 1988, pp. 615-618.

[6] R.H. Heffner et al., "New phase diagram for $(U,Th)Be_{13}$: a muon-spin-resonance and $H_{c1}$ study," *Phys.Rev.Lett.*, vol. 65, 1990, pp. 2816-2819.

[7] C. Geibel et al., "A new heavy-fermion superconductor: $UNi_2Al_3$," *Z.Phys.B - Condensed Matter*, vol. 83, 1991, pp. 305-306.

[8] C. Geibel et al., "Heavy-fermion superconductivity at $T_c = 2$ K in the antiferromagnet $UPd_2Al_3$," *Z.Phys.B - Condensed Matter*, vol. 84, 1991, pp. 1-2.

[9] G. Cordier, G. Dorsam, T. Friedrich, R. Henseleit, and C. Rohr, "$LaAg_3Al_2$, $CeAg_3Al_2$, $PrAg_3Al_2$, $CaAg_{2.2}Al_{2.8}$, $SrAg_{2.5}Al_{2.5}$ and $UNi_2Al_3$: $CaCu_5$ structure type compounds and $CaPd_3Al_5$, a new compound from the $CaCu_5$-derived structure type," *J.Alloys Compd.*, vol. 190, 1993, pp. 201-207.

[10] A. Krimmel et al., "Neutron diffraction study of the heavy fermion superconductors $UM_2Al_3$ (M = Pd, Ni)," *Z.Phys.B - Condensed Matter*, vol. 86, 1992, pp. 161-162.

[11] A. Amato et al., "$\mu^+$SR studies of $UM_2Al_3$, M = Ni,Pd," *Z.Phys.B - Condensed Matter*, vol. 86, 1992, pp. 159-160.

[12] C. Geibel et al., "$UPd_2Al_3$ - a new heavy-fermion superconductor with $T_c$ = 2K," *Physica C*, vol. 185-189, 1991, pp. 2651-2652.

[13] A. de Visser et al., "High-field magnetization of heavy-fermion $UPd_2Al_3$," *Physica B*, vol. 179, 1992, pp. 84-88.

[14] A. Amato et al., "Magnetic and superconducting properties of the heavy-fermion superconductor $UPd_2Al_3$," *Europhys. Lett.*, vol. 19, 1992, pp. 127-133.

[15] E. Brück, Van der Waals-Zeeman Laboratorium, University of Amsterdam, Valckenierstraat, 1018 XE Amsterdam, The Netherlands, personal communication, 1993.

[16] C. Geibel et al., "Magnetism and superconductivity in doped $UPd_2Al_3$ and $UNi_2Al_3$," presented at *Journeés des Actinides*, Bühlerthal, april 1993.

[17] J. Sticht and J. Kübler, "Calculated electronic and magnetic structure of $UNi_2Al_3$ and $UPd_2Al_3$," *Z.Phys.B - Condensed Matter*, vol. 87, 1992, pp. 299-304.

[18] L. Havela, V. Sechovsky, F.R. de Boer, E. Brück and H. Nakotte, "Magnetic anisotropy in UTX compounds," *Physica B*, vol. 177, 1992, pp. 159-163.